

U.S. PAT. NO. 4,912,155, ISSUED MARCH 27, 1990

SERIAL NO.

FILED JUNE 13, 1991

FOR ANTIOXIDANT AROMATIC FLUORO-PHOSPHITES

SUPPLEMENTAL REISSUE PETITION, DECLARATION AND POWER OF ATTORNEY

I, Lester P.J. Burton, residing in New Castle County, State of Delaware, and citizen of Canada, hereby declare that:

1. I believe I am the original, first and sole inventor of the invention entitled ANTIOXIDANT AROMATIC FLUOROPHOSPHITES, described and claimed in the reissue application which was filed on June 13, 1991 and amended through August 25, 1992, and which was originally filed on February 27, 1987 as Application Serial No. 20,023, and which issued as U.S. Pat. No. 4,912,155 on March 27, 1990. Attached to this paper is an Attachment 1 showing a summary of all the amendment to the originally issued claims, and with all the matters in brackets deleted and underlined matter added, constitute the claims sought in this petition.

- 2. I have read and understand the contents of the above-identified specification, including the claims as amended by any amendments referred to above.
- 3. I hereby request that I may be allowed to surrender and do hereby assent to surrender the said U.S. Pat. No. 4,912,155, which is assigned in whole to my former employer, Ethyl Corporation, and request that the Patent may be reissued, upon the foregoing claims.
- 4. I acknowledge the duty to disclose to the Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.
- 5. I believe that the Patent may be wholly or partly inoperative or invalid by reason of my claiming more or less than I had a right to claim in the Patent; and that this was the result of error without any deceptive intention on the part of myself or my employer and assignee of the Patent. The errors relied upon are set forth in the paragraphs below, together with a specification of how and when the errors occurred.
- 6. I was unaware of any errors in the Patent until April 2, 1991, when I met with counsel, and certain errors were identified which are specified below. Other errors were disclosed to me during the course of the prosecution of this reissue application by way of the Examiner rejections.

Claim 1

7. I was assigned the task by my employer to discover new and useful antioxidants for organic materials, mainly polyolefin polymers, especially polypropylene. In the course of my work, I discovered aromatic fluorophosphites that were surprisingly good antioxidants and most surprisingly had far better hydrolytic stability than commercial antioxidants.

- 8. During the development of my invention, I had a librarian conduct a literature search to determine the state of the art. The search uncovered no indications that any aromatic fluorophosphites had ever been disclosed as antioxidants. Several aromatic fluorophosphite chemical structures had been disclosed in the prior art, but none of these were indicated to have any utility to the best of my knowledge. I summarized my results in a memo that on October 8, 1986, I sent to Joseph Odenweller, the attorney then responsible for prosecuting my patent application. That memo is attached as Attachment A.
- 9. I carefully reviewed the specifications and claims of the patent application which Mr. Odenweller prepared. However, in reviewing the claims, I failed to compare the claims against the prior art structures that I had previously uncovered. Having disclosed the structures to the attorney handling the patent application, I simply assumed that the attorney had taken the necessary steps in view of my invention and the prior art. I have limited knowledge of patent law, and relied on the attorney handling the prosecution to make certain that all patent law requirements were met.
- 10. I am now informed that Mr. Odenweller had apparently misunderstood my memo Attachment A. The memo summarized the results of an STN International computerized search. The results provide the structure, and, if the structure appeared only before 1967 there are "0 references" cited, but if the structured appeared in 1967 or after, references are cited. In my memo Attachment A, I had indicated for some structures that there were "0 ref", which meant as indicated above. I am informed that Mr. Odenweller understood that "0 ref" meant that the structure did not appear in the prior art. Unfortunately, I never discussed this memo with any attorney during the prosecution, and, therefore, never explained the import of my notations.

Upon further review of the literature search results, I have also discovered that I overlooked and omitted in my memo certain aromatic fluorophosphite chemical structures that were in the computerized survey. I am now also informed that the attorney inadvertently failed to take into account all the prior art chemical structures that I did supply to him. I believe that these inadvertent failures led to claim 1 of the Patent to potentially claim chemical compounds that were disclosed in the prior art.

- 11. I have now also been shown certain prior art of which I have no recollection of being aware at the time that my original patent application was filed. That art is stated in the Information Disclosure Statement that I understand was filed together with the original reissue petition.
- 12. More specifically, I verily believe that the original issued claim 1 may claim chemical compounds within Formulas V, VI, III or IV which were in the above prior art disclosures, and claim 1 should be further limited by deleting those Formulas V, VI, III and IV. In addition, with regard to claim 1 Formula IV, the Examiner rejected this part of the claim under §112 during his first Office Action of the reissue application. Formula IV was also cancelled in order to avoid this §112 rejection of the claim. I became aware of this problem only after the Examiner rejected this formula during the reissue application.
- 13. In claim 1, the Examiner also noted a typographical error in that "substituent" should have been "substituents". This error arose during the patent application of issued patent 4,912,155. The first time I was aware of the error was after the Examiner rejected the claim during the prosecution of the reissue application.

14. Further, with respect to claim 1, Formula II, the Examiner rejected the claim in view of §112 and requested that the first and second "aryl" be defined and "hydroxy" be deleted from the claim as unsupported in terms of how to make such compounds. I was not aware of these §112 defects in the claim until the Examiner brought them to my attention. The first "aryl" was substituted with "phenyl" which is clearly supported by the specification and examples, and the second "aryl" was also defined as set forth in the specification. In the course of making the above corrections, it was brought to my attention that the punctuation should be corrected in that ":" should be ",". I was not aware that my choice of the terms "aryl" or inclusion of hydroxy" would create §112 problems until the matter was brought to my attention after the filing of the original reissue application. I now see the errors, and wish to have them corrected.

Claims 2-4 and 6

- 15. With regard to claims 2 through 4 and 6 the claims had to be cancelled since they depended upon cancelled formulas in claim 1. The Examiner pointed out during the prosecution that the prior art discloses the compound of claim 2. Therefore, I believe that this claim should be cancelled. I became aware of this problem only after the Examiner rejected this formula during the reissue application.
- 16. Claims 3, 4 and 6, were not rejected over the prior art. These claims were allowable and rewritten in the independent format as claim 43.

Claim 8

17. Claim 8 was amended in view of U.S.S.R. Authorship Certificate 398,574. The amendment makes more clear that the antioxidants of the invention are added to the organic

materials by mixing or spraying and are not substantially reacted with the organic materials, as is shown in the U.S.S.R. Authorship Certificate. I first became aware of the U.S.S.R. Authorship Certificate on April 2, 1991. I now recognize that the use of the term "containing" might potentially cause the claim to read on the reaction of antioxidant with the organic composition, which was not my intent. During prosecution of the reissue application, I also, first became aware that the limitation that the said organic material be a "polymer of an olefinically unsaturated monomer" (the limitation of claim 10) was required to be inserted into claim 8 to avoid a possibility that the prior art (Baranauckus and Spivak) read on claim 8.

Claim 9

18. Claim 9 was amended in view of §112 rejections made for the first time during the prosecution of said reissue application. Claim 9 Formulas I, II and III needed to be corrected for the same reason as stated in paragraph 14, above. The above paragraph 14 is here incorporated by reference. Claim 9 Formula IV was rejected by the Examiner because the specification did not sufficiently demonstrate how to make a compound with the "OH" substituents. I was not aware of that error until the Examiner's rejection was brought to my attention, but now recognize that Formula IV should be deleted. In addition the word "from" was misspelled as "rom". This error occurred during the printing of the patent. There was also another typographical error in that "R₃" should have been "R³" in the first line under Formula III. I first became aware of these errors during the prosecution of the reissue application.

Claim 10

19. Claim 10 had to be cancelled since claim 10 did not further limit amended claim 8. The limitation of claim 10 was inserted into claim 8. Therefore, this claim had to be cancelled.

Claim 11

20. Claim 11 has been amended to depend on newly added claim 44. Newly added claim 44 now restates what had been the "Formula II" portion of original claim 9. The error here is further explained in the discussion of the addition of claims 44 later in this paper. This amendment occurred during the prosecution of the reissue application. I first became of aware of this error during the prosecution of the reissue application.

Claim 12

21. Claim 12 has been amended to depend directly from claim 9, and has been limited to a substituted "R" phenyl group. The errors arose as follows: first, it became apparent during the prosecution that there was no claim directly covering an organic composition with a Formula I compound with substituted phenyl groups, which is one of the preferred embodiments of my invention. Formula I with substituted phenyl groups in the R positions is reflected in the specification. The error of failing to make such composition a specific embodiment first became apparent during the prosecution of the reissue application. The claim had to be changed to be dependent directly on claim 9, rather than claim 11, because the previous limitation that the polymer be of an olefinically unsaturated monomer, which was originally contained in claims 10 and 11, was now incorporated into claim 9, and claim 11 was not made dependent on claim 44.

Claims 13 and 14

22. Claims 13 and 14 have been amended in order to obviate the §112 objections. The first time I became aware of the errors in these claims was during the prosecution of the reissue application. Further the "alkoxycarbonyl" substituents have been deleted to further differentiate the claim from the previously cited Spivak '855 reference. This change was brought about during the prosecution of the reissue application.

Claims 17, 19 and 24

23. Claims 17, 19 and 24 have been amended to obviate the §112 rejection brought to my attention for the first time during the reissue prosecution after the Examiner rejected the claims. The error and the amendments to claim 19 are the same as shown in paragraph 14 above, and paragraph 14 is here incorporated by reference. Further, the word "atoms" was misspelled in the claim issued as "toms", which, I believe was a typographical error in the printing.

Claim 23 and 24

24. Typographical errors in claims 23 and 24 are corrected. The errors were not my fault, but arose in the printing of the original patent by the Patent and Trademark Office. Further, claim 24 contained the same errors as described in paragraph 14 above, and the same amendments were made that are described in paragraph 14. Above paragraph 14 is here incorporated by reference.

Claims 25-31, 41 and 42

25. Claims 25-31, 41 and 42 were rejected over the prior art for the first time during the prosecution of the reissue application. These claims were rejected October 7, 1991, on page

8 of the Office Action. This is the first time I became aware of the error related to these claims.

To expedite prosecution, it was deemed preferable to concede these claims, as opposed to amending the claims to avoid the prior art cited by the Examiner.

Claim 43

26. Claims 43 was added to incorporate the dependent claims 3, 4 and 6 which had to be cancelled since they depended upon cancelled formulas in claim 1 into the independent format. However, these claims were not rejected over the prior art. These claims were allowable and rewritten in the independent format as claim 43. The new claim 43 had to be added to save the otherwise valid claims 3, 4 and 6, which were defective only in that they were dependent from formulas of claim 1 which were overbroad in view of the prior art. The error of claim is detailed in paragraphs 7 through 13, which are incorporated by reference.

Claim 44

27. Newly added claim 44 is essentially of the same scope as was original claim 19 and is of the same scope as the "Formula II" portion of claim 9. Claim 44 had to be added because of errors that occurred in claims 8 and 9, and which are detailed above in paragraphs 17 through 18, which are incorporated by reference. Because of the amendments which had to be made because of errors in claims 8 and 9, as detailed above, the invention of new claim 44, which was covered by the "Formula II" part of originally issued claim 9, was no longer covered by any claim. Therefore, claim 44 had to be added to save the invention which was clearly and specifically disclosed in the specification and which was covered by the originally issued claim 9, Formula II. The first time that it was recognized that claim 44 was needed was when the errors of claims 8 and 9 were disclosed by the Examiner during the present prosecution, and the

original invention of claim 9, Formula II was found to be valid, allowable and no longer covered by the amended claims. Claim 44 does not have the additional limitation of the currently amended claim 8 -- i.e., limiting the "organic material normally susceptible to gradual oxidative degradation" to "being a polymer of an olefinically unsaturated monomer", because claim 8 is inclusive of other antioxidants of, for example, Formulas I and III, while claim 44 is limited to Formula II antioxidants. The limitation had to be included in claim 8 in view of the prior art, but claim 44 is not subject to the same consideration.

- 28. The specification contains corrections to typographical errors in the printed original patent, which errors were not my fault but arose in the printing by the Patent and Trademark Office.
- 29. At Cols. 15 and 16, I have also deleted the disclosure of the alternate use of PBr₃ in making an intermediate in the making of my invention. At the time I originally filed my application I believed that PBr₃ might be a useful reactant. I have since discovered that PBr₃ does not appear to work in the reaction.
- 30. A terminal discloser had to be filed in view of Patent No.4,867,907 which issued prior to Patent No. 4,912,155 (the application involved in the reissue). Patent No. 4,912,155 has an earlier filing date. The Examiner pointed out during the prosecution of the reissue application that double patenting existed. This was the first time I was aware of the double patenting rejection. A terminal disclaimer was filed in order to overcome this rejection.
- 31. As present advised and based on my best recollection, the material listed as offered for sale in the Babullis patent (US Patent No. 4,962,144) at column 3 line 5, was not offered

for sale more than 1 year before the filing of the original application and in fact was offered for

sale after the filing of the patent application of Patent No. 4,912,155.

32. I hereby declare that all statements made herein of my own knowledge are true and

that all statements made on information and belief are believed to be true; and further that these

statements were made with the knowledge that willful false statements and the like so made are

punishable by fine or imprisonment, or both, under 18 U.S.C. 1001 and that such willful false

statements may jeopardize the validity of the application or any patent issued thereon.

33. I hereby appoint the following attorneys or agents to prosecute this application and

to transact all business in the Patent and Trademark Office connected therewith: Rudolf E.

Hutz, Reg. No. 22,397; Thomas M. Meshbesher, Reg. No. 30,982; Robert G. McMorrow,

Reg. No. 30,962; Ashley I. Pezzner, Reg. No. 35,646; Philip M. Pippenger, Reg. No. 25,525

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ATTACHMENT 1

1. An aromatic fluorophosphorus compound suitable for use as an antioxidant said compound being selected from fluorophosphorus compounds having the structure:

Formula V

wherein R is an substituted aryl group wherein the substituents are tert-alkyl groups:

$$(R'O-)_2P-F$$

Formula VI

wherein R' is a substituted aryl group wherein the substituents are selected from sec-alkyl, tertalkyl, aralkyl, cycloalkyl, hydroxy, alkoxy, aryloxy, halo, acyloxy, and alkoxy carbonyl alkyl:]

Formula II

wherein R^1 and R^2 are substituted or unsubstituted [aryl] <u>phenyl</u> groups wherein the [substituent] <u>substituents</u> are selected from alkyl, aryl, aralkyl, cycloalkyl, [hydroxy,] alkoxy, aryloxy, and halo[:], and X is selected from the group consisting of a single bond connecting R^1 and R^2 and divalent bridging groups selected from divalent aliphatic hydrocarbon groups containing 1-12 carbon atoms, -O- and $-S_q-$ wherein q is an integer from 1 to 3[:], and wherein aryl is selected from the group consisting of phenyl, o-tolyl, p-tolyl, naphthyl, 4-phenylphenyl and 4-sec-hexylphenyl.

Formula III

wherein R is a substituted or unsubstituted aryl group wherein the substituents are selected from alkyl, aryl, aralkyl, cycloalkyl, hydroxy, alkoxy, aryloxy, halo, alkoxycarbonyl, alkoxycarbonyl-alkyl and acyloxy, and R³ is selected from the group consisting of alkyl, cycloalkyl, aralkyl, aryl, substituted aryl, alkoxy, cycloalkoxy and aralkoxy; and

wherein R^5 and R^6 are hydrogen or alkyl having 1-12 carbon atoms, y is an integer from 2 to 3, x is an integer from 1 to 3, t is an integer from 2 to 3, u is an integer from 0 to 4 (t+u) equals 2 to 6, w is an integer from 1 to 4, R^7 is hydrogen or an alkyl having 1 to 6 carbon atoms, R^8 is an aliphatic hydrocarbon radical having 1-30 carbon atoms and having valence w, v is an integer from 0-4, R^9 is an aliphatic hydrocarbon radical having 1 to 6 carbon atoms and having valence y.]

- [27. A composition of claim 26 wherein said fluorophosphorus comopund is 2,5-di-tert-butyl-1,4-phenylene bis (difluorophosphite).]
- [28. A composition of claim 26 wherein said fluorophosphorus compound is 4,4'-methylenebis(2,6-di-tert-butylphenyl) bis(difluorophosphite).]
- [29. A composition of claim 26 wherein said fluorophosphite compound is the tris(difluorophosphite ester) of 1,3,5-tris(3,5-di-tert-butyl-4-hydroxybenzyl)-2,4,6-trimethyl benzene.]
- [30. A composition of claim 26 wherein said fluorophosphorus compound is the tetrakis(difluorophosphite ester) of tetrakis(methylene 3-(3,5-di-tert-butyl-4-hydroxyphenyl) propionate)methane.]
- [31. A composition if claim 26 wherein said fluorophosphite compound is difluorophosphite ester of octadecyl 3-(3,5,-di-tert-butylhydroxyphenyl)propioniate.]
- 32. An organic composition of claim 8 further characterized by containing about 0.005-5 wt. percent of a phenolic antioxidant.
- 33. An organic composition of claim 9 further characterized by containing about 0.005-5 wt. percent of a phenolic antioxidant.
- 34. An organic composition of claim 12 further characterized by containing about 0.005-5 wt. percent of a phenolic antioxidant.
- 35. An organic composition of claim 15 further characterized by containing about 0.005-5 wt. percent of a phenolic antioxidant.
- 36. An organic compostion of claim 16 further characterized by containing about 0.005-5 wt. percent of a phenolic antioxidant.

$$(HO-)_s-A (-O-P) = F$$
Formula IV

wherein A is a mono- or poly-nuclear aromatic group, R^4 is independently selected from fluorine, aryloxy, alkylaryloxy, alkoxy and polyalkoxy, r is an integer from 1 to 4, s is an integer from 0 to 3 and (r + s) equals the valence of A.]

- [2. A compound of claim 1 namely bis(2,6-di-tertbutylphenyl) fluorophosphite.]
- [3. A compound of claim 1 namely: bis(2,4-di-tertbutylphenyl) fluorophosphite.]
- [4. A compound of claim 1 namely bis(4-octadecyloxycarbonylethyl-2,6-di-tert-butylphenyl) fluorophosphite.]
- 5. A compound of claim 1 namely: 2,2'ethylidenebis(4,6-di-tert-butylphenyl) fluorophosphite.
- [6. A compound of claim 1 namely: bis-difluorophosphite ester) of 4,4'-methylenebix(2,6-di-tert-butylphenol).]
- 7. A compound of claim 1 namely: 2,2'-bis(4,6-di-tert-butylphenyl) fluorophosphite.
- 8. Organic material normally susceptible to gradual oxidative degradation when in contact with oxygen, said organic material being a polymer of an olefinically unsaturated monomer and having incorporated therein by mixing or spraying [containing] an antioxidant amount of an aromatic fluorophosphorus compound, said compound being characterized by having at least one benzene group bonded through oxygen to a trivalent phosphorus atom and at least one fluorine atom bonded to said phosphorus atom.
- 9. An organic composition of claim 8 wherein said fluorophosphorus compound is selected from the group consisting of compounds having the structures:

$$(RO-)_n P(-F)_{3-n}$$
 Formula I

wherein R is a substituted or unsubstituted [aryl] <u>phenyl</u> group wherein the substituents are selected from alkyl, aryl, aralkyl, cycloalkyl, [hydroxy,] alkoxy, aryloxy, halo, alkoxycarbonyl,

alkoxycarbonylalkyl and acyloxy and n is 1 or 2, and wherein aryl is selected from the group consisting of phenyl, o-tolyl, p-tolyl, naphthyl, 4-phenylphenyl and 4-sec-hexylphenyl;

wherein R^1 and R^2 are substituted or unsubstituted [aryl] <u>phenyl</u> groups wherein the substituents are selected from alkyl, aryl, aralkyl, cycloalkyl, [hydroxy,] alkoxy, aryloxy and halo, and X is selected [rom] <u>from</u> the group consisting of a single bond connecting R^1 and R^2 and divalent bridging groups selected from divalent aliphatic hydrocarbons containing 1-12 carbon atoms, -O- and $-S_q-$ wherein q is an integer from 1 to 3[;], and wherein aryl is selected from the group consisting of pheynl, o-tolyl, p-tolyl, naphthyl, 4-phenylphenyl and 4-sec-hexylphenyl; and

wherein R is as previously defined for Formula I and $[R_3]$ R^3 is selected from the group consisting of alkyl, cycloalkyl, aralkyl, aryl, substituted aryl, alkoxy, cycloalkoxy, aryloxy and aralkoxy[; and], and wherein aryl is selected from the group consisting of phenyl, o-tolyl, p-tolyl, naphthyl, 4-phenylphenyl and 4-sec-hexylphenyl.

wherein A is a mono or polynuclear aromatic group, R^4 is independently selected from fluorine, aryloxy, alkaryloxy, alkoxy and polyalkoxy and r is an integer from 1 to 4, s is an integer from 0 to 3 and (r+s) equals the valence of A].

[10. A composition of claim 8 wherein said organic material is a polymer of an olefinically unsaturated monomer.]

11. A composition of claim [9] 44 wherein said organic material is a polymer of an olefinically unsatruated monomer.



- 12. A composition of claim [11] 9 wherein said compound has Formula I[.], and R is a substituted phenyl group.
- 13. A composition of claim 12 wherein n is 2 and said substituents are selected from alkyls having 1-20 carbon atoms, [aryls having 6-12 carbon atoms] phenyl, o-tolyl, p-tolyl, naphthyl, 4-phenylphenyl, 4-sec-hexylphenyl, aralkyls having 7-12 carbon atoms, cycloalkyls having 5-8 carbon atoms, [hydroxy,] alkoxy having 1-12 carbon atoms, aryloxy having 6-12 carbon atoms, halo, [alkoxycarbonylalkyl having 1-20 carbon atoms in its alkoxy moiety and 1-3 carbon atoms in its alkyl moiety, alkoxycarbonyl having 1-20 carbon atoms in its alkoxy moiety] and acyloxy having 1-4 carbon atoms.
- 14. A composition of claim 13 wherein said substituents are selected from alkyl having 1-20 carbon atoms [and alkoxy carbonylalkyl having 1-20 carbon atoms in its alkoxy moiety and 4 1-3 carbon atoms in its alkyl moiety].
- 15. A composition of claim 14 wherein said fluorophosphite compound is bis(2,6-di-tert-butylphenyl) fluorophosphite.
- 16. A composstion of claim 14 wherein said fluorophosphite is bis(2,4-ditert-butylphenyl) fluorophosphite.
- 17. A composition of claim [14] 12 wherein said fluorophosphite compound is bis(4-octadecyloxycarbonylethyl-2,6-di-tert-butylphenyl) fluorophosphite.
 - 18. A composition of claim 12 wherein n is 1.
- 19. A composition of claim 9 wherein said fluorophosphite compound has Formula II wherein said substituents are selected from alkyl having 1-20 carbon atoms, [aryl having 6-12 carbon atoms,] phenyl, o-tolyl, p-tolyl, naphthyl, 4-phenylphenyl, 4-sec-hexylphenyl, aralkyl having 7-12 carbon atoms, cycloalkyl having 5-8 carbon atoms, [hydroxy,] alkoxy having 1-12 carbon [toms] atoms, aryloxy having 6-12 carbon atoms and halo, and X is selected from the group consisting of a single bond connecting R^1 and R^2 and divalent bridging groups selected from divalent aliphatic hydrocarbon groups containing 1-12 carbon atoms, -O-and -S_a- wherein q is an integer from 1-3.
- 20. A composition of claim 19 wherein said substituent groups are alkyls containing 1-20 carbon atoms.
- 21. A composition of claim 20 wherein said fluorophosphorus compound is 2,2'-ethylidenebis(4,6-di-tert-butylphenyl) fluorophosphite.



- 22. A composition of claim 20 wherein said fluorophosphorus compound is 2,2'-methylenebis (4-methyl-6-tert-butylphenyl) fluorophosphite.
- 23. A composition of claim 20 wherein said fluorophosphite compound is [22,2,] 2.2'-bis(4,6-di-tert-butylphenyl) fluorophosphite.
- 24. A composition of claim 9 wherin said fluorophosphorus compound has Formula III wherein said substituents are selected from alkyls having 1-20 carbon atoms, [aryls having 6-12 carbon atoms,] phenyl, o-tolyl, p-tolyl, naphthyl, 4-phenylphenyl, 4-sechexylphenyl, aralkyls having 7-12 carbon atoms, cycloalkyls having 5-8 carbon atoms, [hydroxy,] alkoxy having 1-12 carbon atoms, aryloxy having 6-12 carbon atoms, halo, alkoxycarbonylalkyl having 1-20 carbon atoms in its alkoxy moiety and 1-3 carbon atoms in its alkyl moiety, alkoxycarbonyl having 1-20 carbon atoms in its alkoxy moiety and acyloxy having 1-4 carbon atoms, and R³ is selected from alkyl having 1-20 carbon atoms, cycloalkyl having 5-8 carbon atoms and aralkyls having 7-12 carbon atoms which are bonded through [oxyqen] oxygen to phosphorus and aryls having 6-12 carbon atoms, alkyl having 1-20 carbon atoms, cycloalkyls having 5-8 carbon atoms and aralkyls having 7-12 carbon atoms which are bonded directly to said phosphorus.
- [25. A composition of claim 9 wherein said fluorophosphorus compound has Formula IV.]
 - [26. A composition of claim 25 whrerein A has a structure selected from:

Structure IV (i)

Structure IV (ii)

$$R^3$$
 R^6 R^6 R^6

Structure IV (iii)

Structure IV (iv)

Structure IV (vi)

$$R^{\delta} \left(\begin{array}{c} O \\ \parallel \\ -C-C_{r}H_{2r} - \end{array} \right) \left[\begin{array}{c} R^{\delta} \\ R^{\delta} \end{array} \right]$$

Structure IV (vii)

Serial No. 07/714,441

AN 5585/RE

- 37. An organic composition of claim 17 further characterized by containing about 0.005-5 wt. percent of a phenolic antioxidant.
- 38. An organic composition of claim 19 further characterized by containing 0.005-5 wt. percent of a phenolic antioxidant.
- 39. An organic composition of claim 21 further characterized by containing 0.005-5 wt. percent of a phenolic antioxidant.
- 40. An organic composition of claim 39 wherein said phenolic antioxidant is 1,3,5-tris(3,5-di-tert-butyl-b 4-hydroxybenzyl)-2,4,6-trimethylbenzene.
- [41. An organic composition of claim 39 further characterized by containing about 0.005-5 wt. percent of a phenolic antioxidant.]
- [42. An organic composition of claim 25 further characterized by containing about 0.005 -5 wt. percent of a phenolic antioxidant.]
- --43. A aromatic fluorophosphorus compound suitable for use as an antioxidant, said compound being selected from the group consisting of bis(2,4-di-tert-butylphenyl) fluorophosphite; bis(4-octadecyloxycarbonylethyl-2,6-di-tert-butylphenyl) fluorophosphite; and 4,4'-methylenebis(2,6-di-tert-butylphenyl)bis (difluorophosphite).--
- --44. A compound of claim 1 combined in an antioxidant amount with an organic material normally susceptible to gradual oxidative degradation when in contact with oxygen.--

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Attachment A to
Declaration of Lester P.J. Burton

ETHYL CORPORATION

INTER-OFFICE

TO KA Keblys

Address F7C

FROM LPT Buton

SUBJECT Fluorophosphites

DATE 12/10/85

A computer search of the substructure PhOPF was executed. The search yielded 171 structures and 52 references The majority of the hits were metal complexes, phosphates or pentaroardinate species. No reference to antioxidant activity was found. The arylfluorophosphites found are listed below:

CC RL Shulkin Ic willnesk Ar,PF

(RR'N) P (NHR' could be in place of RR'N). Named in connection with the antistatic treatment of plastics. 144

s-Hexamethylene-diaminophosphite. Named in connection with the production of profiled plastic strips which are weather and light stable. 119

O,O-diethyl N,N-diethylphosphoroamidite. Used for the preparation of pesticides. ""

Amides and imides of phosphorous acid. Catalyze the polymerization of formaldehyde to high molecular weight poly(oxymethylenes). 127

Polycondensates from PCl₃ and such as hexamethylene diamine. Applicable as ion exchangers.

 $(R_2N)_3P$, R = Me, Et, Pr, Bu, especially Me. Improve leaded gasoline with regard to preignition and octane number.

Di-Et N-(2,4-diMe-phenyl) phosphoramidite. Is an additive for motor gasoline, improving the octane number, and minimizing combustion zone deposits without lowering the octane number. 241

"N,N',N"-(trioctylphenyl)phosphorous triamide" "Di-Bu-Nphenyl amidophosphite." Named as antiknock additives for gasoline. 289

PhOP(NCO)₂. Named in connection with pigmented polyurethane coating compositions having improved viscosity stability.

I. LIST OF COMPOUNDS

I.1. Phosphites

I.1.1. Difluorophosphites

TYPE: ROPF 2

CH₃OPF₂. ^{31}P -111 ppm, J_{PF} 1275 Hz. $^{62}8$ $F_{2}POCH_{2}CH_{2}OPF_{2}$. $(CH_{2}OP)_{2}Cl_{4}$ + SbF₃. b_{100} 50°, n_{D}^{26} 1.3523, ^{19}F NMR, 1249 ^{31}P -112.0 ppm, J_{PF} 1295 Hz. 1209 ,

Propf₂. Ropcl₂ + Sbf₃. b. 44.5°, n_D² 1.3400, ¹⁹F
 NMR, ¹²⁴⁹ ³¹P -111.5 ppm, J_{PF} 1287 Hz. ¹²⁰⁹, ¹²⁴⁹
CH₂:CHCH₂OPf₂. ROPCl₂ + Sbf₃. b. 42°, ¹⁹F NMR, ¹²⁴⁹
³¹P -111.9 ppm, J_{PF} 1290 Hz. ¹²⁰⁹, ¹²⁴⁹
BuOPf₂. ROPCl₂ + Sbf₃. b. 75°, n_D² 1.3580, ¹²⁴⁹ ³¹P
 -111.9 ppm, J_{PF} 1288 Hz. ¹²⁰⁹
PhOPf₂. PhOPCl₂ + Sbf₃. b₆ 58°, n_D²⁷ 1.4575, ¹⁹F
 NMR, ¹²⁴⁹ ³¹P -110.1 ppm, J_{PF} 1326 Hz. ¹²⁰⁹, ¹²⁴⁹
1,4-C₆H₄(OPf₂)₂. -PCl₂ + Sbf₃. b₁₂ 59°, n_D²³ 1.4488, ¹⁹F NMR, ¹²⁴⁹ ³¹P -109.8 ppm, J_{PF} 1328 Hz. ¹²⁰⁹, ¹²⁴⁹

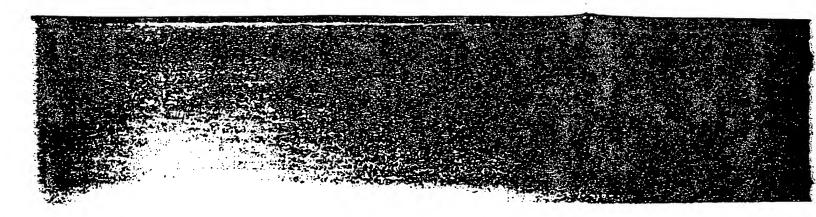
I.1.2. Monofluore

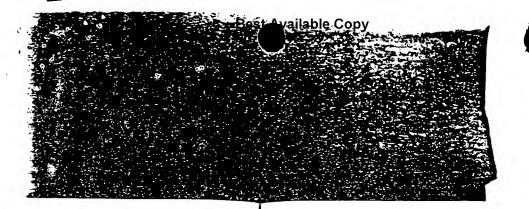
OCH₂CH₂OPF. (RO)₂PC1 b₁₈ 26°, ¹²⁰¹ d₄²³ 1.4039, MR_D 19.90 -124.4 ppm, JpF 17 ÓCH (Me) CH₂OÞF. (RO) 2 ! ngº 1.4035, MRD 24 OCH (Me) CHMeOPF. (RO): n_D^{20} 1.4020, MR_D 29 OCH (Me) CH 2 CH 2 OPF. (RC n_D^{20} 1.4160, MR_D 29 OCH2.C(Et) (Bu)CH2OPF. 1.1241, n_D 1.4765 NMR. 1261 OCH₂ (CH₂)₂CH₂OPF. (RO n_D² 1.4450, MR_D 30 OCH₂ (CH₂), CH₂OPF. (RO n_D^{20} 1.4270, MR_D 39 OCH 2 (CH 2) 8 CH 2 OPF. (RC n_D^{20} 1.4798, MRD 57 $1,2-C_{6}H_{10}O_{2}PF$. (RO) ₂F n_{D}^{20} 1.4586, MR_D 36 1,2-C₆H₄O₂PF. (RO)₂PC b₆ 36.5°, 12 4 9 b₆ 3 n_D, 1.5080, 12 4 9 n_D 19 F NMR, 12 4 9 31 p JPOCCH ca. 1 Hz, J $3-Me-1, 2-C_6H_3O_2PF.$ (R n_D^2 1.5170, MR_D 39 n_D²⁰ 1.5170, m_{RD} 34-Me-1,2-C₆H₃O₂PF. (F n_D²⁰ 1.5220, MR_D 39 1,2- $\dot{C}_{6}H_{4}C$ (:0) $\dot{O}_{7}F_{1}$ (RC \dot{n}_{D}^{25} 1.5390. 1249

I.1.3. Dichloroph

TYPE: ROPCl2

CD₃OPCl₂. Ib. b₆₀ 31 MeOPCl₂. Ia. b₇₅₈ 95 1.47725,^{299,740} 31 C6H5CH(CO2Et)OPCl2. $1.2720, n_0^{1} 1.5259$ (-)-mandelate α_D^{16}





phorous Acid

of RR'N). Named in coneatment of plastics. 15 Named in connection d plastic strips which

idite. Used for the

.cid. Catalyze the to high molecular

as hexamethylene changers.248 ially Me. Improve preignition and octane

amidite. Is an addiwing the octane number, deposits without lower-

us triamide" "Di-Bu-Nis antiknock additives

ith pigmented polyureing improved viscosity

Z. 628. 1. b₁₈₀ 50°, n_D²⁶) ppm, JpF 1295 Hz. 1209,

 n_{D}^{20} 1.3400, 19 F 287 Hz. 1209 , 1249 19F NMR, 1249 42°,) 1.3580, 1249 31P

n_D²⁷ 1.4575, ¹⁹F 326 Hz. ¹²⁰⁹, 1249 2 59°, n_D²³ 1.4488, F 1328 Hz. 1209,1249 I.1.2. Monofluorophosphites with P in Ring System

OCH₂CH₂OPF. (RO)₂PCl + SbF₃. 1064,1201,1249 b₁₇₀ 48°, 1249 b₁₈ 26°, 1201 d₄0 1.3552, 1201 n₀3.5 1.4003, 1249 n₀0 1.4039, MR_D 19.90 (20.56), 1201 19F NMR, 1249 31P -124.4 ppm, J_{PF} 1223 Hz, 1209,1249 1H NMR. 432

Me)CH₂OPF. (RO)₂PCl + SbF₃. b₁₀₀, 44°, d²⁰₄ 1.2226, n_0^{20} 1.4035, MR_D 24.78 (25.18), n_0^{1064} , n_0^{1201} IR. n_0^{1201} OCH (Me) CH 2 OPF.

OCH (Me) CHMeOPF. (RO) $_{2}$ PCl + SbF $_{3}$. b $_{16}$ 28°, d $_{4}^{20}$ 1.1568, n_{D}^{20} 1.4020, MRD 29.08 (29.79). $_{1201}^{1201}$

OCH (Me) CH_2CH_2OPF . (RO) $_2PC1 + SbF_3$. b_{16} 37°, d_4^2 ° 1.1857, n_D^2 ° 1.4160, MR_D 29.22 (29.79). 1201

OCH₂.C(Et) (Bu) CH₂OPF. (RO)₂PCl + SbF₃. b₁ 61° (d² ° 1.1241, n² ° 1.4765, MR_D 52.28 (52.88), 1201 (H

OCH₂ (CH₂)₂CH₂OPF. (RO)₂PCl + SbF₃. b₁₆ 38°, d²⁰₄ 1.2180, n_D^{20} 1.4450, MR_D 30.16 (29.80), 1201 IR, 1201 H NMR. 1201

OCH₂ (CH₂) $_{8}$ CH₂OPF. (RO) $_{2}$ PCl + SbF₃. b₂ 80°, d₄° 1.1041, n₀° 1.4798, MR_D 57.09 (57.50). 1201

nố 1.4798, MRD 57.09 (57.50). 1201

1,2-C₆H₁₀O₂PF. (RO)₂PCl + SbF₃. b₁ 34°, d₄° 1.2140,

n_D° 1.4586, MR_D 36.93 (36.83), 1201 1H NMR. 1201

1,2-C₆H₄O₂PF. (RO)₂PCl + SbF₃, 1201, 1249 or + NaF. 1249

b₆ 36.5°, 1249 b₆ 38°, 1201 d₄ 1.3592, 1201 n_D° 1.5092,

n_D° 1.5080, 1249 n_D° 1.5160, MR_D 35.13 (35.43), 1201

19F NMR, 1249 31P -123.1 ppm, J_{PF} 1305 Hz, 1H NMR, 1201

JPOCCH ca. 1 Hz, JPOCCCH < 0.5 Hz, 1209, 1249

3-Me-1, 2-C₆H₃O₂PF. (RO)₂PCl + SbF₃. b₂ 58°, d₄° 1.3045,

n_D° 1.5170, MR_D 39.94 (calc. 40.04). 1202

4-Me-1, 2-C₆H₃O₂PF. (RO)₂PCl + SbF₃. b₇ 84°, d₄° 1.3150,

n_D° 1.5220, MR_D 39.92 (calc. 40.04). 1202

 $1.2-\dot{C}_{6}H_{4}C(:0)O\dot{P}F_{1}$, $(RO)_{2}PC1 + KSO_{2}F_{1}$ $b_{0.15-0.2}$ $44-7^{\circ}$, n_{D}^{25} $1.5390.^{124.9}$

I.1.3. Dichlorophosphites

TYPE: ROPCl₂

CD₃OPCl₂. Ib. b₆₀ 31-2°, d²⁴ 1.3892, n_0^2 1.4682. n_0^3 MeOPCl₂. Ia. b₇₅₃ 95-6°, d² 1.4275, d²⁰ 1.3980, n_0^3 1.47725, n_0^2 1.47725, n_0^2 1.47725, n_0^2 1.4725, n_0^2 1.4725, n_0^2 1.4725, n_0^2 1.5259, n_0^2 1.2827, d²¹ 1.2720, n_0^2 1.5259, n_0^2 1.17.5° (1 = 10 cm) from the (-)-mandelate n_0^2 131.0°.

Schwabe, Sandmair, Marx PO Box 86 02 45 8000 Munchen 86

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Patent & Trademark Division
Attn: Ms. Patricia J. Hogan
451 Florida Boulevard
Baton Rouge, Louisiana 70801
U.S.A.

DIPLICHEM, DR. JUR DR RER NAT KURT SANDMAR DIPLIPHYS, DR. RER NAT LOTHAR MARX

DIPL. ING. HANS-GEOPG SCHWABE

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STUNTZSTRASSE 16 8000 Munchen 80

July 12, 1988

Ini Zeichen Your ref Votre ref

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European Patent Application No. 88 101 945.9

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Yours sincerely,

On First Sandmair

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| DiplIng Dr. Dr. S | . Schwabe, andmair, | • | | | |
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| | | | <u> </u> | "Datum/Date | |
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| Zeicher/Ret/Rét | | Anmeldung Nr./Application | n No./Demande | nº//Patent Nr./Patent No./Brevet nº | |
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EUROPEAN SEARCH REPORT

EP 88 10 1945

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| | The present search report has I | peen drawn up for all claims | | |
| т | Place of search HE HAGUE | Date of completion of the se 03-06-1988 | | Examer SLIER L.M. |
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| SU-A- 393574 | | None | | |
| US-A- 3254050 | | . None | | |
| US-A- 3281506 | | None | | |
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| Полимер | про | WEIIII | при р Прогрев | рушающего на- растяжении в ева пленок при | | |
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| Исходный полимер Полимер, обработан- пый дихлор-(пента- метилфенил) фосфи- | 425 | 173 | 0 | 0 | 0 . | |
| том Полимер, обработан- ный дихлорфенилфос- | 430 | 430 | 462 | 415 | 262 | |
| фитом | 575 | 160 | 0 | 0 | 0 | |

В качестве аминов могут быть использованы пиридин, пиперидин, анилин и другие

кипятят еще 2 часа. После охлаждения до комнатной температуры высаживают полимер добавлением 250 мл метилового спирта. Осадок отфильтровывают, промывают многократ-5 но ацетоном и сущат в вакуумном шкафу при температуре 80-100°С и остаточном даплении 100 мм рт. ст. Выход составил 95%.

Предмет изобретения

Способ стабилизации полифениленовых эфиров путем взаимодействия их с фосфорсодержащими стабилизаторами в присутствии аминов или алкоголятов щелочных металлов, отличающийся тем, что, с целью повышения устойчивости полифениленовых эфиров к возленствию высоких температур, в качестве фос-

$$R_{1} \xrightarrow{R_{1}} R_{1} = 0 - P < R_{1} + H_{2} = 0$$

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$$R_{2} \xrightarrow{R_{3}} R_{4} = 0 - P - 0$$

$$R_{3} \xrightarrow{R_{1}} R_{4} = 0 - P - 0$$

где $R_1 = C_1 - C_{10}$ -алкилы;

$$R = Hal; \quad -0 \xrightarrow{R_1 R_1} R_1$$

$$R = R_1 R_1$$

Hal = F, Cl, Br, J,

Me = Li, Na, K, Rb, Cs

алифатические или ароматические амины. Реакшию лучше проводить при избытке галонд- 20 соединен и обессы формулы фосфита.

Следующий пример иллюстрирует данное изобретение.

Пример 1. К раствору 10,0 г поли-2,6-диметил-1,4-фениленоксила (xapakteристическая вязкость 0,75 в бензоле при 25°C) в 130 мл толуола добавляют раствор метилата натрия, полученный добавлением 0.1 г натрия и 5,0 мл метилового спирта. Реакционную смесь кипятят 1 час, добавляют избыток дихлор (пентаметилфенил) фосфита (2,8 г) и

форсодержавых слабализаторов используют

$$R-P = \begin{cases} R_1 & R_1 \\ 0 - \sum_{R_1 = R_1}^{R_1} R_1 \end{cases}$$

25 rge R=Hal:
$$-0 - \underbrace{R_1 R_1}_{R_1 R_1}$$

30 $R_1 = C_1 - C_{10}$ -ankii m_i : $ile^i = F_i$ Cl, B_{f_i} J

Составитель В. Поляков

Редактор Л. Емельянова Техред Л. Богданова Корректоры Л. Царькова и О. Тюрица

Энкэл № 1289 : Изд. № 1062 : 11 7 7 7 7 2 2 65

..... ЦИНИЛИ Государственного комитета Совета Министров СССР ³ по делам изобретений и открытий,

Москва, Ж-35, Pavmeras наб. 1/5

мот, Засорский филипл

DECLARATION, POWER OF ATTORNEY, AND PETITION

Case No. 5585

| As: | a below | named | inventor, | I | hereby | declare | that: |
|-----|---------|-------|-----------|---|--------|---------|-------|
| | | | | | | | |

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:_

ANTIOXIDANT AROMATIC FLUOROPHOSPHITES

the specification of which is attached hereto.

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37,

Code of Federal Regulations, Section 1.56(a). I hereby claim the benefit under Title 35, United States Code, Section 120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, Section 112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, Section 1.56(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

| | FILING DATE | STATUS | | |
|------------------------|-------------|--------|--|--|
| APPLICATION SERIAL NO. | FILING DATE | | | |
| NONE | | | | |
| | | | | |
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And I hereby appoint Donald L. Johnson, Reg. No. 17,076; John F. Sieberth, Reg. No. 17,704; Joseph D. Odenweller, Reg. No. 22,361

All of Ethyl Corporation, 451 Florida Boulevard, Baton Rouge, Louisiana 70801; and Arthur G. Connolly, Reg. No. 13,416 of Farmers Bank Building, Tenth and Market Streets, Wilmington, Delaware 19899; or any one of them my attorneys or agents to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith, said Donald L. Johnson and John F. Sieberth and either one of them to have full power of substitution and revocation, including the power to revoke the appointments of attorneys or agents herein at (504) 388-8188 made. Please make all telephone calls to Joseph D. Odenweller

Address all correspondence to: Mr. John F. Sieberth, Patent & Trademark Division, Ethyl Corporation, 451 Florida Boulevard, Baton Rouge, Louisiana 70801.

WHEREFORE, I pray that Letters Patent be granted to me or us for the invention or discovery described and claimed in the foregoing specification and claims, and I hereby subscribe my name to the foregoing specification and claims, declaration, power of attorney, and this

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief petition. are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

| | | | DATE |
|---|----------------------|-------------|-------------|
| FULL NAME OF SOLE OR FIRST JOINT INVENTOR | INVENTORS SIGNATURE | (5) | Feb 24,1987 |
| LESTER P. J. BURTON | West 1 | CITIZENSHIP | |
| RESIDENCE | T. 70917 | Canada | |
| 17321 Monitor Avenue, Baton Rou | ige, LA /USI/ | Canada | |
| POST OFFICE ADDRESS 17321 Monitor Avenue, Baton Rou | | | |
| | INVENTOR'S SIGNATURE | | DATE |
| FUEL NAME OF SECOND JOINT INVENTOR, IF ANY | | | * |
| | | CITIZENSHIP | |
| RESIDENCE | | | |
| | | <u> </u> | |
| POST OFFICE ADDRESS | _ | | |
| | | | DATE |
| FULL NAME OF THIRD JOINT INVENTOR, IF ANY | INVENTOR'S STONATURE | | |
| | | CITIZENSHIP | <u></u> |
| RESIDENCE | | | |
| | | | |

24

| 6 7 8 | $\mathbb{R}^9 \left[\begin{array}{c} \\ \\ \\ \\ \\ \\ \\ \\ \end{array} \right]_{Y}$ | Structure IV (ii) |
|----------------------------|--|--------------------|
| 9 10 11 | s_x R^5 R^6 R^6 | Structure IV (iii) |
| 12 13 14 | \mathbb{R}^5 \mathbb{R}^6 \mathbb{R}^6 | Structure IV (iv) |
| 15 16 17 18 19 | (R ⁷ -) _u CH ₂ R ₆ | Structure IV (v) |
| 20 21 22 23 | $_{R}^{8}$ ($_{C}$ $_{C}$ $_{C}$ $_{V}^{H}$ $_{2v}$ | Structure IV (vi) |





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| 07/714441 | 06/13/91 | Burton | 1 | | | |
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| | | | | | ART UNIT | PAPER NUMBER |
| | | - | | | 1202 | 241/2 |
| | | | | DAT | E MAILED: | |
| | EX | AMINER INTERV | VIEW SUMMAR | Y RECORD | | |
| All participants (applicant, | applicant's representative | e, PTO personnel): | | | | |
| (1) A shiey | I. Pezzn | er | (3) | | | |
| (2) | Shah | | _ (4) | | | |
| Date of interview | 6/10/93 | | - | | | |
| Type: 🔲 Telephonic | Personal (copy is given | to Dapplicant | Napolicant's repr | esentative) | | |
| | | | | | | |
| Exhibit shown or demonstr | ation conducted: LI Yo | es [M. No. If yes, I | brief description: _ | | | |
| Agreement | _ | • | question. | s not reached. | | |
| Claims discussed: | As of rea | cord | | | | |
| Identification of prior art d | iscussed: A | ream | | | | |
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| Description of the general r | nature of what was agreed | I to if an agreement v | vas reached, or any | other comment | s() Stampe | d Post card |
| bory has see | n privided f | w the de | claration | (2) The | term " | rzanic materi |
| will be change | ed to "a po | lymer q | an olefin | ically u | nsaturate | d monomer" |
| 3) Claim 44 | will be che | exed for i | porsiste i | ntenjer | ence witz | 4,962,144 |
| 5) support for | Markush l | s) recessor | been prodifi | ided or Cations | in other | 4,962,144, lines 22-25 claims as |
| (A fuller description, if ne attached. Also, where no co | cessary, and a copy of t | he amendments, if a | vailable, which the | e examiner agree | ed would render the | claims allowable must be |
| Unless the paragraphs belo NOT WAIVED AND MUS last Office action has alread | w have been checked to T INCLUDE THE SUBS | indicate to the cont TANCE OF THE IN | rary, A FORMAL TERVIEW (e.g., it | WRITTEN RES | SPONSE TO THE Le reverse side of this | AST OFFICE ACTION IS form). If a response to the |

Mukund J. Shah

response requirements of the last Office action.

☐ Since the examiner's interview summary above (including any attachments) reflects a complete response to each of the objections, rejections and requirements that may be present in the last Office action, and since the claims are now allowable, this completed form is considered to fulfill the

 \square It is not necessary for applicant to provide a separate record of the substance of the interview.